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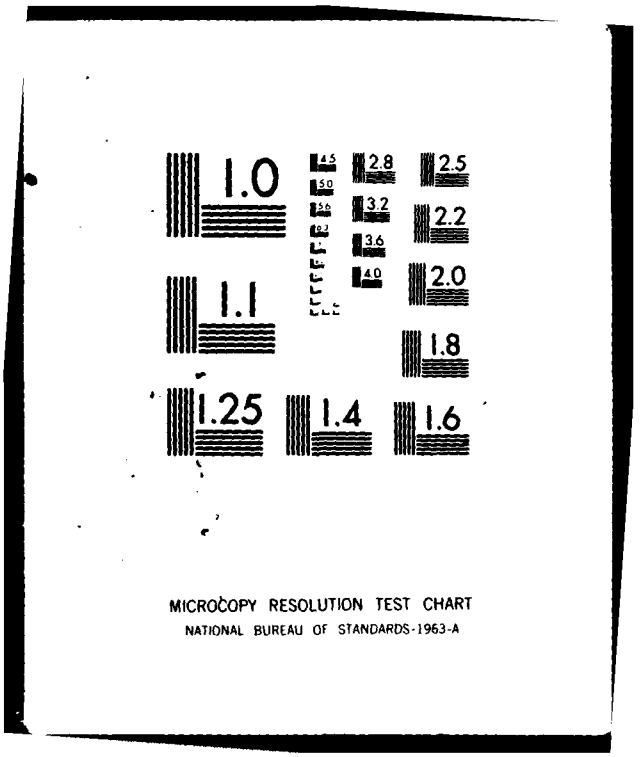

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High-Energy Positron Ionization of Adsorbed Species in the Impulse Approximation

by

Kai-Shue Lam and Thomas F. George

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University of Rochester
Department of Chemistry
Rochester, New York 14627

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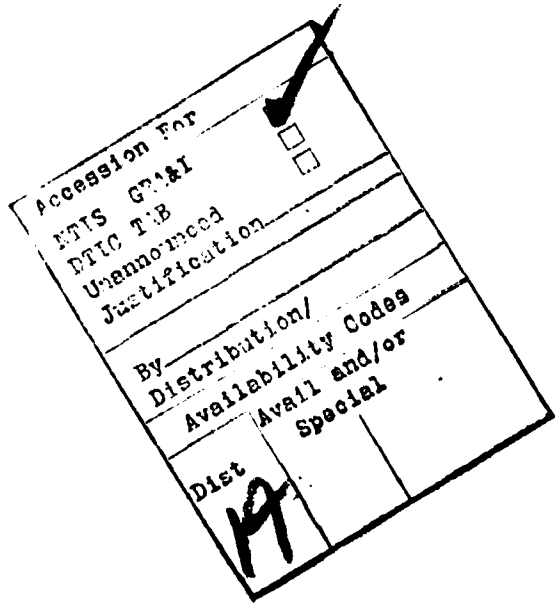
Abstract. High-Energy Positron Ionization of adsorbed species is treated using the one-electron group-orbital linear-chain model and within the impulse approximation. Inter-orbital interference effects are found to be significant, leading to quenching (towards the low impact energy regime) and enhancement (at high impact energies) of ionization compared to positron gas-phase ionization.



I. Introduction

Recently, positron sources (available beam energy up to ~400 eV) have come to be of value in the diffraction studies of solid surfaces.[1] These studies complement the traditional low-energy electron diffraction (LEED) ones and point to new directions in the use of positrons as excitation or ionizing agents in the study of chemisorption bond-characters. Unlike electrons, positrons lead to no exchange effects during collision and thus allow for simplifications in theoretical analyses, provided the effects of positronium formation can be neglected (which should be a good approximation for beam energies of several hundred eV). Furthermore, in ionizing situations, the distinguishability of the product particles (positrons and electrons) also facilitates the measurement of ionization cross-sections.

Here we report on a theoretical treatment of high-energy positron ionization of adsorbed species based on the one-electron group-orbital linear-chain (OEGOLC) model of the adatom-surface system (with only nearest-neighbor interactions),[2,3] and the impulse approximation (IA)[4] for the description of the collision process. Indirect effects of the solid substrate on the ionization process, such as electron- and positron-phonon interactions, positron diffusion within the solid, positronium formation and positron annihilation, etc. will be ignored. Such effects are either expected to be weak compared to direct positron-electron interaction, or are negligible under the conditions of the high impact energies considered here (50-400 eV). Our main purpose is to investigate the effects of orbital mixing in the chemisorption bond on ionization. Hence we will focus on systems with

a localized adatom-surface (chemisorption) bond (corresponding to a localized state with energy lying outside a band). In this case, if the surface coverage is large enough, the ejected electrons will be mainly from near the surface, i.e., from the localized orbitals, and these electrons would contribute predominantly to the ionization cross-section compared to the "secondary" non-localized electrons. In our model, the positron is accordingly assumed to interact only with electrons in the localized bound state. The OEGOLC model, though somewhat qualitative, is the simplest one capable of producing a localized state. The IA, on the other hand, has been applied with considerable success to complex collision events such as high-energy gas-phase collisional ionization.[5] Our calculations indicate that inter-orbital interference effects in the group orbital picture are significant in the ionization process: there is a slight quenching towards the low energy regime (<90 eV) and considerable enhancement at high energies, compared to gas-phase ionization.

II. Theory

(a) The ionization cross-section

The physical process considered here is the collision between an incident positron (with beam energy considerably in excess of any bound-electron energies) and an adatom, and the subsequent single-ionization of the adatom. The IA assumes that, during collision, the interaction between the bound electrons of the adatom-surface system is "turned off". This implies that the incident positron can only spend a very short time in the field of the bound electrons. The electron interacting directly with the positron is "momentarily"

free and has a momentum ($\vec{k}$) distribution given by the Fourier transform $\tilde{\psi}_0(\vec{k})$ of the one-electron chemisorption wave function. The positron-electron system is then described by a positive-energy Coulomb wave function[6] weighted by $\tilde{\psi}_0(\vec{k})$. If $\vec{r}_1$ is the positron coordinate, and $\vec{r}_2$ and $\vec{r}_3$ are coordinates of the electrons in the localized state, the positron-electron system wave function in the IA is thus given by (in what follows, all quantities will be expressed in terms of atomic units)

$$\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3) = \frac{1}{\sqrt{2}}\{\psi_0(\vec{r}_2)\int d^3k_3\tilde{\psi}_0(\vec{k}_3)\psi_{\vec{k}_1,\vec{k}_3}(\vec{r}_1,\vec{r}_3) + \psi_0(\vec{r}_3)\int d^3k_2\tilde{\psi}_0(\vec{k}_2)\psi_{\vec{k}_1,\vec{k}_2}(\vec{r}_1,\vec{r}_2)\}. \quad (1)$$

In (1), $\vec{k}_1$ is the wave vector of the incident positron, $\psi_0(\vec{r})$ is the one-electron chemisorption wave function of the localized state, $\tilde{\psi}_0(\vec{k})$ is its Fourier transform, referred to above, and $\psi_{\vec{k}_i,\vec{k}_j}(\vec{r}_i,\vec{r}_j)$ is the Coulomb wave function for a positron-electron pair given by (on factoring into CM and relative coordinates)

$$\psi_{\vec{k}_1,\vec{k}_2}(\vec{r}_1,\vec{r}_2) = \frac{1}{(2\pi)^3}\exp\{\tfrac{1}{2}i(\vec{k}_1+\vec{k}_2)\cdot(\vec{r}_1+\vec{r}_2)\}\psi_{\frac{1}{2}(\vec{k}_1-\vec{k}_1)}(\vec{r}_1-\vec{r}_2) \quad (2)$$

where

$$\psi_{\vec{k}}(\vec{r}) = e^{\pi/2k}\Gamma(1-\tfrac{i}{k})e^{i\vec{k}\cdot\vec{r}}\,{}_1F_1(\tfrac{i}{k};1;ikr-i\vec{k}\cdot\vec{r}). \quad (3)$$

$\psi_{\vec{k}}(\vec{r})$ is the wave function in the continuous spectrum for an attractive Coulomb potential; ${}_1F_1$ is the confluent hypergeometric function.

The final states of the positron and ejected electron can be approximated by plane waves, and the final-state wave function for the positron-electron system can be written as

$$u_f(\vec{r}_1,\vec{r}_2,\vec{r}_3) = \frac{1}{\sqrt{2}} \frac{e^{i\vec{k}'\cdot\vec{r}_1}}{(2\pi)^3}\{e^{i\vec{k}_2'\cdot\vec{r}_2}\psi_0(\vec{r}_3) + e^{i\vec{k}_2'\cdot\vec{r}_3}\psi_0(r_2)\}, \tag{4}$$

where $\vec{k}_1'$ and $\vec{k}_2'$ are the wave vectors for the scattered positron and emitted electron, respectively. The single-ionization scattering amplitude f is then given by

$$f = -\frac{1}{\sqrt{2\pi}}(T_2+T_3), \tag{5}$$

where T_i, the transition matrix elements, can be expressed as

$$T_i = \int d^3r_1 d^3r_2 d^3r_3 u_f^*(\vec{r}_1,\vec{r}_2,\vec{r}_3)(-\frac{1}{r_{1i}})\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3); \tag{6}$$

and $r_{ij} = |\vec{r}_i - \vec{r}_j|$. Using (1) and (4) in (6) and expanding, we obtain a sum of eight terms for f, each of which represents different initial and final configurations for the positron-electron system coupled by a distinct $1/r_{ij}$ Coulomb force. Of these, in the Born approximation, four vanish and four are equal to one another, say, T. Based on this observation, we introduce the approximation that

$$T_2 + T_3 = 4T \tag{7}$$

where T is still calculated within the IA:

$$T = -(\tfrac{1}{2})\frac{1}{(2\pi)^3}\int d^3r_1 d^3r_2 e^{-i\vec{k}_1'\cdot\vec{r}_1 - i\vec{k}_2'\cdot\vec{r}_2}\frac{1}{r_{12}}\int d^3k_2 \tilde{\psi}_0(\vec{k}_2)\psi_{\vec{k}_1,\vec{k}_2}(\vec{r}_1,\vec{r}_2). \tag{8}$$

Using (2) in (8), we obtain

$$T_2 + T_3 = -\frac{2}{(2\pi)^3}\tilde{\psi}_0(\vec{k}_1'+\vec{k}_2'-\vec{k}_1)\int d^3r \frac{e^{-i(\frac{\vec{k}_1'-\vec{k}_2'}{2})\cdot\vec{r}}}{r}\psi_{\frac{1}{2}(2\vec{k}_1-\vec{k}_1'-\vec{k}_2')}(\vec{r}). \tag{9}$$

The integral in (9) has been evaluated by several authors,[7] and gives

$$T_2 + T_3 = -\frac{4}{(2\pi)^2}\tilde{\psi}_0(\vec{k}_1'+\vec{k}_2'-\vec{k}_1)\exp(\pi/|2\vec{k}_1-\vec{k}_1'-\vec{k}_2'|)$$
$$\times \frac{\Gamma(1-2i/|2\vec{k}_1-\vec{k}_1'-\vec{k}_2'|)}{(\vec{k}_1-\vec{k}_1')^2}\{e^{-i\pi}\frac{(\vec{k}_1-\vec{k}_1')\cdot(\vec{k}_1-\vec{k}_2')}{(\vec{k}_1-\vec{k}_1')^2}\}^{-2i/|2\vec{k}_1-\vec{k}_1'-\vec{k}_2'|} \tag{10}$$

The wave vectors are required to satisfy the condition of energy conservation: $\kappa^2 = k_1^2 - 2|\varepsilon| = k_1'^2 + k_2'^2$, where $-|\varepsilon|$ is the bound energy of the localized electron. The total single-ionization cross section σ is then given by

$$\sigma = \frac{1}{k_1}\int_{k_2'<\kappa} d^3k_2' d\Omega_1', k_1' |f(\vec{k}_1', \vec{k}_2')|^2. \tag{11}$$

In (11), $d\Omega_1'$ designates the directions of the scattered positron. Only "backward-scattered" directions of $\vec{k}_2'$ need be considered since these are the only observable ones. The scattering amplitude used in (11) is then given by

$$|f(\vec{k}_1', \vec{k}_2')|^2 = \frac{2^4(2\pi)^2 |\tilde{\psi}_0(\vec{k}_1'+\vec{k}_2'-\vec{k}_1)|^2 \exp(-2\pi/|2\vec{k}_1-\vec{k}_1'-\vec{k}_2'|)}{|2\vec{k}_1-\vec{k}_1'-\vec{k}_2'||\vec{k}_1-\vec{k}_1'|^4 \sinh(2\pi/|2\vec{k}_1-\vec{k}_1'-\vec{k}_2'|)} \tag{12}$$

(b) The chemisorption wave function

The chemisorption wave function in the OEGOLC model can be written, using the LCAO approximation, as

$$\psi_0(\vec{r}) = C_\lambda \phi_a(\vec{r}) + \sum_{m=0}^{N-1} C_m \phi(\vec{r}+(\lambda+m)a\hat{z}), \tag{13}$$

where $\hat{z}$ is in the direction of the chain (outward from the surface), a is the inter-atomic distance in the linear chain, λa is the distance between the adatom and the first substrate atom, N+1 is the total number of substrate atoms in the chain, and $\vec{r}$ is measured from the nucleus of the adatom. We assume that only two types of orbitals, ϕ_a (corresponding to the free adatom) and ϕ (corresponding to individual substrate atoms) enter into ψ_0. [This assumption should be reasonable for most alkalis and some transition metals.] The coefficients C_λ and C_m depend on the following parameters:

ε_a (bound energy corresponding to ϕ_a), α (bound energy corresponding to ϕ), β' (interaction between the adatom and the first substrate atom) and β (nearest-neighbor interaction between the substrate atoms).

When (13) is used in the Schrödinger equation with a one-electron Hamiltonian, the vanishing of the secular determinant for the coefficients C_λ and C_m leads to the following result. A localized state (ψ_0 damped in the crystal) exists if the equation

$$(\cos\theta + \sin\theta \cot N\theta)\left(\frac{\alpha-\varepsilon_a}{\beta} + 2\cos\theta\right) = \left(\frac{\beta'}{\beta}\right)^2 \tag{14}$$

has at least one complex solution for θ, ($\theta = i\xi$ or $\pi+i\xi$, $\xi>0$ and real), with energy given by $\varepsilon = \alpha \pm 2\beta \cosh\xi$. $|\tilde{\psi}_0|^2$ in (12) is then given by

$$|\tilde{\psi}_0(\vec{k})|^2 = C_\lambda^2\tilde{\phi}_a^2 + \tilde{\phi}^2[1-C_\lambda^2+2\sum_{m'>m} C_{m'}C_m\cos\{a\vec{k}\cdot\hat{z}(m'-m)\}]$$
$$+ 2C_\lambda\tilde{\phi}_a\tilde{\phi}\sum_m C_m\cos\{(\lambda+m)a\vec{k}\cdot\hat{z}\}, \tag{15}$$

where $\tilde{\phi}_a$ and $\tilde{\phi}$ are the Fourier transforms of ϕ_a and ϕ, respectively, and both are taken to be real. The secular equations for C_λ and C_m also lead to

$$C_\lambda = \left\{\frac{\beta'/\beta}{(\alpha-\varepsilon_a)/\beta \pm 2\cosh\xi}\right\}C_0 \tag{16a}$$

and

$$C_m = \frac{\sinh\{(N-m)\xi\}}{\sinh N\xi} C_0 \tag{16b}$$

For a localized (damped wave function, the series in (15) terminates quickly. (15) displays clearly the inter-orbital interference effects.

III. Results and Discussion

Calculations have been carried out for the H(adatom)/Li(substrate) system with normally incident positrons. β and β' are estimated by the Wolfsberg-Helmholz formula,[8] and the overlap integrals required therein are obtained from tabulations from Mulliken et al.[9] ϕ_a and ϕ are taken to be the H 1s and Li 2s Slater AO's, respectively. The following parameters are then obtained (in a.u.): $\beta = -0.17$, $\beta' = -0.35$, $\varepsilon = -0.76$. Table 1 gives the normalized coefficients C_λ and C_m for N = 100 computed using (16). To the accuracy stated, these results are identical to those for N = 14. The energy gap between the localized state and the bottom edge of the 2s band is 0.22 a.u.

The integrations in (11) are carried out numerically using the 6- and 16-point Gaussian quadratures, and the ionization cross sections are compared with those for the positron ionization of the H atom, also computed using the IA (Fig. 1). In the latter case, only a simple orbital enters into (12), and inter-orbital interference effects are absent. [For purposes of comparison, σ for the H atom case only include "backward" ionization directions.] Our results indicate that at high impact energies there is a distinct enhancement in ionization due to the interference effects of the group orbital, while at low energies these effects tend to quench the ionization. The peak cross section, however, occurs at a higher energy for the adatom case than for the gas-phase case. Even though the low energy results are less reliable due to the doubtful validity of the IA in this regime, the increasing probability of positronium formation, and hence positron annihilation,

also tend to decrease the ionization cross section. Hence the overall trend is expected to be quenching in the low-energy regime and enhancement in the high energy regime. The enhancement may be attributed to the fact that, at high energies, the positron probes deeper into the crystal.

TABLE 1. Normalized coefficients for the localized H/Li chemisorption wavefunction with bound energy $\epsilon = -0.76$ a.u.

m	m=λ	0	1	2	3	4	5	6	7	8
C_m	0.8005	0.5637	0.1917	0.0648	0.0220	0.0073	0.0025	0.0009	0.0003	0

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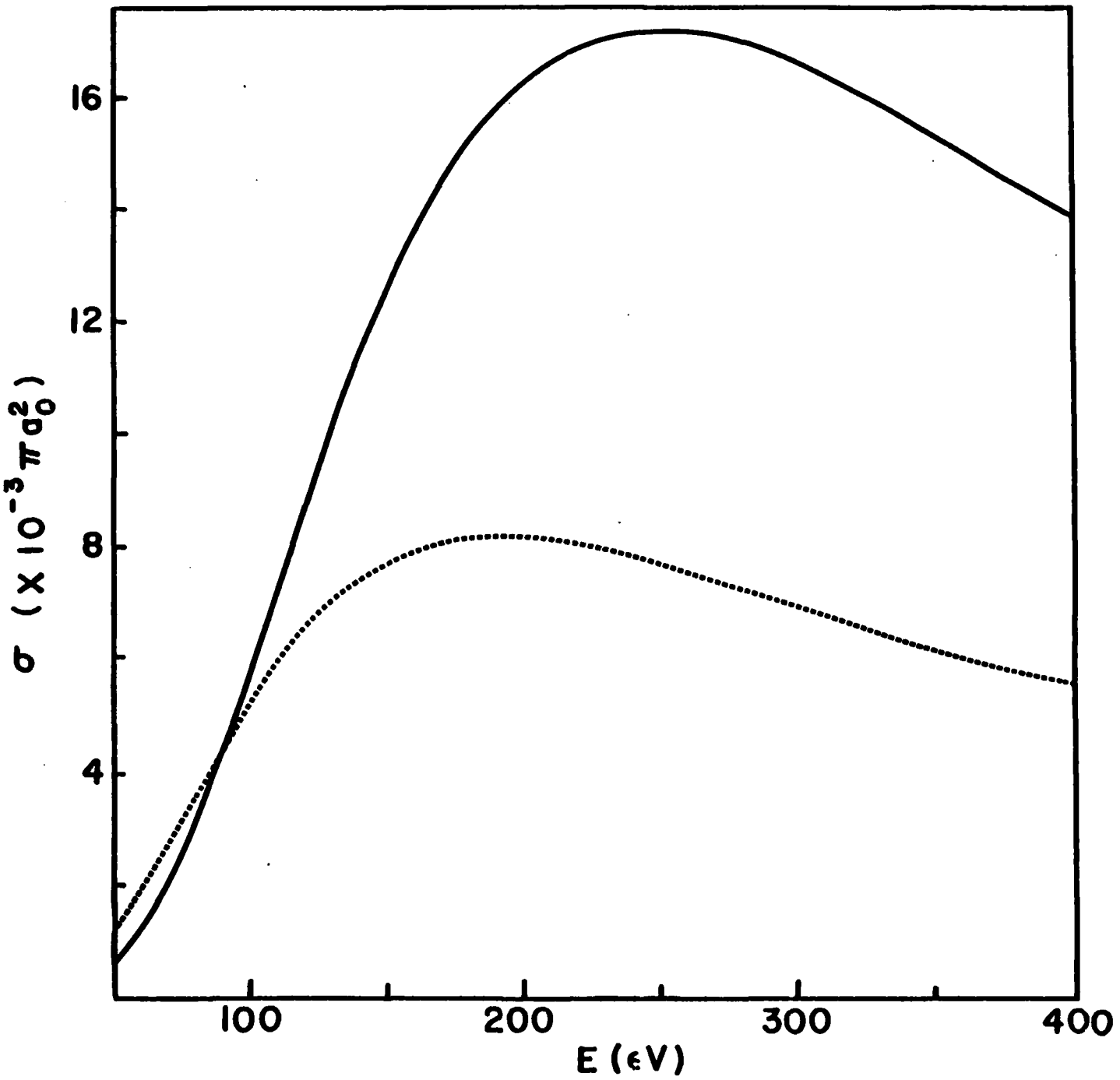
Fig. 1. Positron single-ionization cross sections (σ) for H/Li and H atom as a function of incident positron energy (E). a_0 = 1 Bohr. ———, H/Li; -----, H atom. In both cases only "backward" ionization directions are included.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σ (X 10⁻³ πa₀²)
100
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E (eV)

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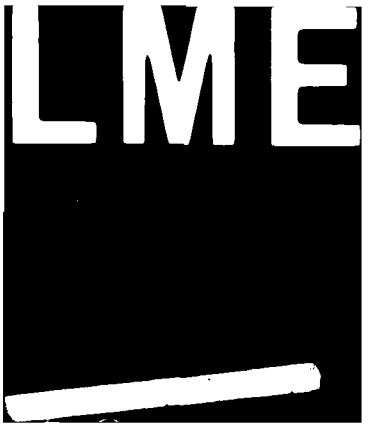
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